

## **New advances in engineered backfill materials: phosphate as a potentially important add-on to the existing bentonite concept.**

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### **ABSTRACT:**

The aim of this proposal is to advance the functionality of engineered barrier systems (EBS) by investigating how the addition of phosphate minerals (phosphates) would increase sorption/uptake efficiency of radionuclides by backfill material in the scenario of water breaching the repository at saturated conditions. To study immobilization mechanisms for iodine-129 ( $^{129}\text{I}$ , a mobile fission product) and uranium ( $U$ , a primary component of nuclear spent fuel), we propose to investigate the  $I$  and  $U$  entrapment by phosphates from aqueous solution at a range of temperatures from 300°C (thermal peak of the waste package) to 25°C (standard state).

Studies on radionuclides and structural information for phosphates, together with our preliminary results, strongly indicate that  $U$  and its mobile fission products can be efficiently immobilized through uptake from aqueous solution by phosphates. With the aim to develop a new engineered backfill material, we propose to investigate the mechanisms of such uptake at a range of temperatures, pH, and redox conditions. Available data on the uptake of  $I$  and  $U$  are often qualitative and mostly restricted to low temperatures (~25°C) where adsorption of radionuclides at the mineral surface (often a reversible process) is prevalent. At high temperatures, dissolution/crystallization processes occur much faster than at low temperatures and therefore, the uptake of radionuclides by their incorporation and burying in the phosphate lattice is expected. Phosphates not only effectively entrap  $U$ , but also retain  $U$  and its decay products for millions of years. Therefore, the addition of phosphates to the backfill material is expected to significantly enhance immobilization of  $U$  and its decay products in EBS. Here we propose to quantify the entrapment of  $I$  and  $U$  by phosphates and evaluate the uptake mechanisms at variable pH, temperature, and redox state. To achieve the above goals, we will employ a number of experimental techniques, which include benchtop and hydrothermal crystallization experiments and post experimental characterization with: X-ray diffraction (XRD), scanning electron microscopy (SEM), electron backscattered diffraction (EBSD), inductively coupled plasma mass spectrometry (ICP-MS), laser ablation (LA) ICP-MS, electron microprobe analyzer (EMPA), ultraviolet–visible (UV-Vis) spectrophotometry, atom probe tomography (APT), and synchrotron-based X-ray spectroscopy (XAS) and micro X-ray fluorescence ( $\mu$ -XRF) methods. Expected results will provide novel quantitative information on the interaction of spent nuclear fuel with aqueous solutions at different pH, redox conditions, and temperatures (elevated from radioactive decay) as an immediate consequence of ground water breaking through EBS. Obtained data will permit us to optimize the composition of a new backfill mixture for improved effectiveness. In addition, the proposed research will improve the current knowledge of the following mineralogical and geochemical aspects critical to nuclear environmental remediation: 1) The mechanisms of  $I$  and  $U$  incorporation into mineral phases and quantitative constants permitting to evaluate this process under a range of relevant conditions; 2) The role of  $I$  and  $U$  incorporation in formation of associated mineral phases. Proposed collaboration is strongly beneficial for both execution of the project and for study on bentonite-phosphate interaction, which is being performed in EES-14 at LANL.



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